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SELF-PHASE MODULATION: A REVIEW

January 1975

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Self-phase modulation is described from an experimental and theoretical point of view. Applications for the self-phase modulated continua are given. Recommendations are made for future studies.

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INTRODUCTION

In the following paragraphs self-phase modulation (SPM) will be reviewed. A brief background will be given and the process will be described from a theoretical and experimental point of view. The materials studied and the results of the studies will be discussed. The applications of this technique will be explored along with the appropriate experiments. Several future experiments will be proposed.

BACKGROUND

When a highly intense light pulse propagates through a medium, the electric field of the pulse can alter the index of refraction, which in turn, affects the propagation of the pulse. The time dependent index changes the phase of the wave which changes the temporal and spectral characteristics as well. The spectral broadening which occurs is called self-phase modulation or super broadening. There are a number of mechanisms which can contribute to the index variation: (1) direct electronic cloud distortion, (2) molecular reorientation, (3) molecular redistribution, and (4) librational motion of molecules.

Spectral broadening was first observed in CS_2 by Brewer[1] in 1967. A Q-switched ruby laser with 100 kW peak power was self-focused in the CS_2. The spectral composition of the self-trapped filaments was observed to not correspond to Raman, Rayleigh, or Brillouin scattering. These experiments were repeated in CS_2 by Shimizu[2] who observed a regular periodic structure superimposed on the frequency broadened spectrum. These results were interpreted in terms of a molecular orientational model. Polloni[3] et al., studied self-focusing and self-phase modulation in CS_2 and interpreted their results in terms of a librational model in which the molecules "rock" in the field of the neighboring molecules.

Brewer and Lee[4] used a mode locked 1.06μm laser to study self-trapping in low viscosity liquids, high viscosity liquids and glass where linear and rotational diffusion modes are frozen out. Their results support a molecular electronic distortion mechanism for the index of refraction change. The importance of the electronic dis-

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[1] R. G. Brewer, Phys. Rev. Lett. 19, 8 (1967).

[2] F. Shimizu, Phys. Rev. Lett. 19, 1097 (1967).

[3] R. Polloni, C. A. Sacchi, and O. Svelto, Phys. Rev. Lett. 23, 690 (1969).

[4] R. G. Brewer and C. H. Lee, Phys. Rev. Lett. 21, 267 (1968).

tortion mechanism is shown also in several papers by Alfano and Shapiro[5-8]. Self-phase modulation has been investigated recently both experimentally and theoretically near the electronic resonances of a PrF_3 crystal.[9] Information was obtained on the evolution of the SPM spectrum through and beyond the electronic resonances.

THEORETICAL

The problem of an intense electromagnetic wave traveling in a medium where the index of refraction depends on the electric field of the light has been studied.[8-14] There are a variety of effects which can occur under the self-action of light waves. Some of these effects are self-phase modulation, self-focusing, self-defocusing, and self-steepening. The following analysis pertains mostly to self-phase modulation:

In looking at picosecond spectra from various media one finds that the light is often self-focused into filaments. This can occur in materials which are homogeneous in the absence of the electromagnetic

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[5] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. 24, 1217 (1970).

[6] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. 24, 592 (1970).

[7] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. 24, 584 (1970).

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[14] F. DeMartini, C. H. Townes, T. K. Gustafson, and P. L. Kelley, Phys. Rev. Lett. 164, 312 (1967).

wave if the dielectric constant increases with field. A phenomenological explanation[12] can be obtained by considering a uniform circular beam of diameter (d) propagating through a medium in which the index of refraction is:

$$n = n_o + n_2E^2 + \ldots \tag{1}$$

Ordinarily, when all but the first term in the expansion is neglected, the beam would be expected to expand with angular divergence $\theta \sim 1.22\lambda/n_o d$. However, if the term n_2E^2 is large enough the critical angle for total reflection at the beam edge will be such that all of the energy is confined to the beam. In other words spreading of the beam by diffraction does not occur. The power at which this occurs is:

$$P = \frac{\pi d^2}{4}\frac{n_o E^2 c}{8\pi} \geq (1.22\lambda)^2 \frac{c}{64 n_2} \quad \text{for } \theta << 1. \tag{2}$$

Equation (1) can be rewritten as follows in a time dependent form for a single index affecting mechanism of relaxation time τ.

$$n(t) = n_o + \frac{n_2}{\tau}\int_{-oo}^{t} e^{-(t-t')/\tau} E^2(t')dt' \tag{3}$$

The electric field of the incident laser pulse is:

$$E(t) = E_o e^{-t^2/T^2} \cos(\omega_o t), \tag{4}$$

where T is related to the pulse width at half-maximum. By writing the equation in the above form we have neglected chirping.[15] As the laser pulse propagates through the medium the electric field changes in phase as follows:

$$E(t) = E_o e^{-t^2/T^2}\left[\cos \omega_o t - \phi(t)\right] \tag{5}$$

where

$$\phi(t) = n(t)\omega_o z/C\,. \tag{6}$$

This approach ignores dispersion. Dispersion broadens the pulse in time and moves the longer wavelengths to the leading edge of the pulse.

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12 R.Y. Chiao, E. Garmire, and C.H. Townes, Phys. Rev. Lett. 13, 479 (1964).

15 E.B. Treacy, Phys. Lett. 28A, 34 (1968).

The spectral density of the light which is modulated in phase is given by

$$S(\omega) = (c/4\pi)\,|E(\omega)|^2 \qquad (7)$$

where $E(\omega)$ is the Fourier transform of $E(t)$. Therefore, it can be seen that the spectral density depends on the electric field of the laser pulse in the medium, and the electric field of the laser pulse in the medium depends on how the medium responds to the electric field of the incident laser pulse. To give an example, consider that the n_2E_2 term arises solely from an electronic mechanism which has a relaxation time $\tau < 1/\omega_o$. In other words the index of refraction responds at optical frequencies. In this case the function $(1/\tau)\exp\left[-(t - t')/\tau\right]$ in Equation 3 may be replaced by $\delta(t - t')$ and Equation 3 becomes

$$n(t) = n_o + n_2 E_o^2 \exp(-2t^2/T^2) \cos^2(\omega_o t), \qquad (8)$$

and Equation 5 becomes

$$E(t) = E_o \exp(-t^2/T^2)\cos\left[\omega_o t - \omega_o n_o z/c - n_2 E_o^2 \omega_o z/c \exp(-2t^2/T^2) \times \cos^2(\omega_o t)\right] \qquad (9)$$

Alfano et al.,[8] Fourier transformed Equation 9, and the spectra for the electronic mechanism were computed numerically for different values of $n_2E_o^2\omega_o z/c = \beta$ and T_f which is the pulse full width at half-maximum times $\sqrt{2}$. The spectra are symmetric around the input frequency and have a nonperiodic structure of maxima and minima throughout. For $\beta = 120$ and $T_f = .4$ psec the spectra extend over 14000 cm^{-1}. Going to longer pulses markedly reduces the spectral content.

Using the approach of Shimizu[2] the instantantaneous frequency shift may be defined as $\partial\,\phi(t)/\partial t$. This leads to the following relationship

$$\Delta\omega = \omega_o z n_2 E^2/c\tau. \qquad (10)$$

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[2] F. Shimizu, Phys. Rev. Lett. 19, 1097 (1967).

[8] R. R. Alfano, L. L. Hope, and S. L. Shapiro, Phys. Rev. A. 6. 433 (1972).

Substituting $\omega_o \sim 2 \times 10^4$ cm^{-1}, $z \sim 1$ cm, $c \sim 3 \times 10^{10}$ cm/sec, $n_o E_o^2 \sim 10^{-4}$ esu and $\tau \sim 1 \times 10^{-14}$sec, $\Delta\omega \sim 2/3 \times 10^4$cm^{-1}. This is the amount of broadening expected from an electronic mechanism. From the above equation it would appear that much slower effects (τ longer) would have a smaller frequency spread (sweep). However, the value of n_2 for the Kerr effect associated with molecular reorientation is about 10^3 times greater than the electronic Kerr effect. Therefore, one can expect a substantial amount of broadening from these slower ($\sim$10 psec) processes associated with molecular reorientation.

EXPERIMENTAL

A typical experimental arrangement for generating self-phase modulated light is usually comprised of a mode locked Nd or ruby laser, a frequency doubler, a lens for focusing the laser light into a sample cell and a lens for imaging the filaments onto the slits of a spectrograph.

Alfano and Shapiro[5] have demonstrated that significant changes in atomic shape occur under the intense fields of picosecond light pulses in liquid argon and liquid and solid krypton. They focused 5300Å mode-locked laser light into a 12 cm sample cell. The beam intensity in the medium was $\sim 4 \times 10^{11}$W/cm^2. Spectral sweeps of 1000-6000 cm^{-1} were observed on both sides of the laser light. This experiment demonstrated that the electronic mechanism can contribute substantially to SPM.

Frequency broadening was observed in calcite, quartz, sodium chloride, and glass[6] and the mechanism was identified as electronic self-phase modulation. For these studies a Q-switched Nd-glass mode-locked system was used with a KDP doubler. The pulse width was 4 psec and the power was $\sim 2 \times 10^8$ W at 5300Å. The beam was reduced in size by an inverted telescope to 1.2 mm diameter. The filament size in the samples $\sim 20\mu$. Typical sweeps were as follows:

MATERIAL	CRYSTAL LENGTH	STOKES SWEEP	ANTI-STOKES SWEEP
Calcite	4 cm	4400 cm^{-1}	6100 cm^{-1}
Quartz	4.5 cm	3900 cm^{-1}	5500 cm^{-1}
Sodium Chloride	4.7 cm	3900 cm^{-1}	7300 cm^{-1}
Dense Flint Glass	7.55 cm	1100 cm^{-1}	-------
Borosilicate Crown	8.9 cm	4200 cm^{-1}	7400 cm^{-1}
Light Barium Crown	8.9 cm	4200 cm^{-1}	7400 cm^{-1}

[5] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. 24, 1217 (1970).

[6] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. 24, 592 (1970).

It was also noted in this paper the SPM was observable within the KDP doubler. The large amount of spectral broadening in these materials indicates relaxation times $\sim 10^{-15}$sec, therefore these materials might be useful as a ultrafast light gate.[16]

SPM has been observed in H_2O and D_2O,[17,18] The continua generated in these media were recorded by a 3/4 meter spex spectrograph. The Stokes continua was cut-off by the red response of the Polaroid Type 47 film. The anti-Stokes emission extends beyond 3100Å. Sharp inverse Raman absorptions were noted in both media. A measurement of the time width of the continuum pulse shows it to be at least as short as the incident laser pulse.

SPM has been observed in CS_2 by many authors.[1,2,11,19] It was observed that the broadening occurred in discrete bands of frequencies extending to either side of the laser frequency. The bands may be several to tens in number, with a total frequency spread of tens to hundreds of wavenumbers. Cheung et al.,[11] proposed a model which fit their data very well. The model assumes two plane waves of slightly different frequency traveling in a nonlinear medium. The spectral extent of the continua from CS_2, as well as the degree of symmetry between the Stokes and anti-Stokes light, and the band spacing depend on the time and frequency characteristics of the incident pulse.

The Stokes and anti-Stokes spectra from PrF_3 were investigated using an intense 5300Å picosecong pulse.[9] The peak power was $\sim 8 \times 10^8$ W and the pulse width was $\sim$ 4 psec. PrF_3 was chosen for this study because of the nearness of the energy of the electronic levels to the energy of the laser light. The continuum extends past the absorptions of the Pr^{3+} ion; >3000 cm^{-1} on the Stokes side (film limit) and >6000 cm^{-1} on the anti-Stokes side. The intensity of the continuum outside the absorptions is

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[1] R. G. Brewer, Phys. Rev. Lett. 19, 8 (1967).

[2] F. Shimizu, Phys. Rev. Lett. 19, 1097 (1967).

[9] R. R. Alfano, J. I. Gersten, G. A. Zawadzkas, and N. Tzoar, Phys. Rev. A. 10, 698 (1974).

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[17] G. E. Busch, R. P. Jones, and P. M. Rentzepis, Chem. Phys. Lett. 18, 178 (1973).

[18] W. Werncke, A. Lau, M. Pfeiffer, K. Lenz, H. J. Weigmann and C. D. Thuy, Opt. Comm. 4, 413 (1972).

[19] J. Reintjes, R. L. Carman, and F. Shimizu, Phys. Rev. A. 8, 1486 (1973).

$\sim 10^{-4}$ of the intensity of the central maximum, and the intensity of the continuum inside the absorptions is $\sim 10^{-2}$ of the intensity of the central maximum.

Additional theoretical and experimental evidence was given supporting the direct electronic distortion model of SPM in calcite, liquid CCL_4, liquid argon, solid krypton, and liquid nitrogen.[8] The spectra were characterized by large spectral widths and nonperiodic substructure with an occasional interference structure. The spectral extent on each side of the laser line was the same within 20 percent. The Stokes and anti-Stokes intensities were approximately the same and 10^2 - 10^3 less intense than the central frequency.

SPM was observed in CS_2, toluene, benzene and nitrobenzene by using a 6 cm cell, a 10 cm focusing lens, a 3/4 m spectrograph and hypersensitized Kodak I-Z spectroscopic plates.[19] The spectra showed interference patterns with the intensity and spacing of the maxima increasing with distance from the central maxima. The average spacing between peaks is $\sim$ 50 cm^{-1}. The extent of the broading was found to vary considerably from filament to filament. The largest broadening was found in CS_2 with spectra extending 500 - 1000 cm^{-1} on the long and short wavelength sides. The maximum broadening on both sides for toluene was 300 - 500 cm^{-1}, and for benzene and nitrobenzene was 200 - 300 cm^{-1}. The broadening was found to be fairly symmetrical. The ratio of the energy in the Stokes side to the anti-Stokes side was estimated to be between 1 and 2. The degree of symmetry found here is much greater than that found in spectra generated by Q-switched multi-mode lasers. Although, differences are also found in the spectra generated by different mode-locked lasers.

APPLICATIONS

The super-broadened continua arising from self-phase modulation is an extremely valuable research tool because of the frequency extent and the short time frame of the light. A variety of experiments have been performed utilizing these characteristics, and in the following paragraphs a few examples will be given.

[8] R. R. Alfano, L. L. Hope, and S. L. Shapiro, Phys. Rev. A. 6, 433 (1972).

[19] J. Reintjes, R. L. Carman, and F. Shimizu, Phys. Rev. A. 8, 1486 (1973).

The inverse Raman spectra has been observed in benzene, carbon disulfide, methanol, liquid nitrogen, and calcite using a 4 psec self-phase modulated continuum.[20] Picosecond pulses at 5300Å were focused into borosilicate BK-7 glass producing light from 4000Å to 7000Å. The low dispersion allows the 5300Å light and the continuum to emerge from the glass simultaneously. The continuum and the laser pulse then enter the sample and are subsequently imaged onto film in a spectrograph. The result is a photograph of the continuum with the Raman absorptions superimposed.

In a recent paper a picosecond flash photolysis and spectroscopy experiment was performed on 3,3'-diethyloxadicarbocyanine iodide.[21] In In this experiment the 5300Å light and the continuum are separated and delayed by various amounts up to 4 nsec. One of the interesting features of this experiment is that only the central region of the sample was pumped by the 5300Å pulse and the continuum passed through the entire sample. Therefore, each spectrum has the excited state absorption imaged between the normal absorption, i.e., each spectral shot has its own reference. By introducing suitable delays in the continuum arrival the time dependence of the excited states was established. The excited singlet lifetime is $1.15 \pm .15$ nsec in ethanol and 560 ± 70 psec in water.

The transient absorption spectra of DTTC was studied using picosecond ruby pulses and a super-broadened continua over the range 400 - 900 nm.[22] A transient absorption band centered at 525 nm with 35 nm width was observed. The dynamical behavior of this absorption was studied utilizing an echelon technique. This echelon delays different segments of the continuum by 13 psec with a total range of 210 spec. The ruby pulse is then timed to be coincident with a given segment of the continuum at the sample. Because segments of the continuum are spread out spatially as well the spectrograph shows the absorption spectra for each segment. In this way the time and frequency characteristics of the transient absorption can be established in a single shot. In this case it was determined that the band at 525 nm has a lifetime of 90 ± 30 psec.

FUTURE STUDIES

SPM will be studied in a variety of materials with large electro-optic coefficients which are useful as Q-switches. These materials all have large electric field dependent indices of refraction, with long

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[20] R. R. Alfano and S. L. Shapiro, Chem. Phys. Lett. 8, 631 (1971).

[21] D. Magde and M. Windsor, Chem. Phys. Lett. 27, 31 (1974).

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relaxation times. Also, those materials with a reasonably accessible ferroelectric to paraelectric transition will be studied for SPM in the paraelectric state near the transition temperature. If the optical field in these materials can create a local ferroelectric state there will be a substantial change in the index of refraction and a concomitant increase in the SPM. The high fields developed over the short times should allow the examination of the induced polar state in a time and electric field regime never before studied.

In addition to the materials with large electro-optic coefficients a variety of molecules will be studied which are similar but are different in symmetry. For example, various nitrobenzenes and toluences will be compared as well as carbon tetrachloride with chloromethane.

All of the above experiments are designed to aid in the understanding of the SPM process. It is only through the understanding of this effect that it can be either minimized when it is operating as a loss mechanism in high powered lasers or maximized when a highly intense, broad band, short pulsewidth source of radiation is needed. It will be particularly interesting to explore methods for maximizing the SPM in each of the above experiments.

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